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Wim J. van Ooij

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**IN THE UNITED STATES PATENT & TRADEMARK OFFICE**

Applicant: van Ooij et al.

Paper No.:

Serial No.: 09/356,926

Group Art Unit: 1775

Filing Date: July 19, 1999

Examiner: Michael LaVilla

For: **SILANE COATINGS FOR METAL**

**DECLARATION UNDER 37 C.F.R. 1.132**

BOX Patent Application  
Assistant Commissioner for Patents  
Washington, DC 20231

Dear Sir:

Dr. Wim J. van Ooij declares that:

1. He is a co-inventor of, and is familiar with the present U.S. Patent Application Serial No. 09/356,926, filed July 19, 1999, entitled "Silane Coatings for Metal." He is also familiar with the Official Actions issued in this application, including the Official Action dated March 19, 2002 and the prior art references cited therein.

2. The methods of treating a metal surface as defined by claim 1 of the present application are based upon the surprising discovery that the addition of one or more bis-silyl aminosilanes to a vinyl silane solution significantly improves the storage stability of the solution and significantly improves the corrosion protection provided by the solution. As demonstrated in the examples on pages 16-18 of the specification, untreated HDG panels exhibited significant corrosion, while the addition of even a small amount of bis-

(trimethoxysilylpropyl) amine ("A-1170") to a vinyltrimethoxsilane solution provided astonishing improvements in corrosion protection.

3. In order to demonstrate that bis-silyl aminosilanes other than A-1170 provide these same unexpected results, the following additional experiments were carried out under his direction and control:

### Experiment 1

Silane solutions were prepared at 5% for vinyltriethoxysilane (VS), bis-(trimethoxysilylpropyl) amine ("A-1170") and bis-(trimethoxysilylpropyl) ethylene diamine ("Y9400"). The structure of Y9400 (available from OSi Specialties) is shown on page 10 of the specification. A solvent (ethanol) was used to improve silane solubility, and the ratio of the silane/deionized water/ethanol was 5/5/90 by volume. The solutions were stirred at room temperature in order to hydrolyze the silanes. A first silane solution was mixed at the ratio of 9/1 of VS/Y9400 and a second silane solution was mixed at the ratio of 9/1 of VS/A-1170. The pH of the resulting mixtures VS/Y9400 and VS/A-1170 were both 7.5. The pH of the 5% vinyl silane solution was 6. Alkaline cleaned hot-dip galvanized steel panels (HDG, available from ACT Laboratories) were treated using the VS/Y9400, VS/A-1170 mixtures and the 5% vinyl silane solution and then cured at 100 °C for 30 minutes.

The corrosion performance of the silane-treated HDG panels were evaluated using a salt immersion test. The electrolyte was a 3.5% neutral NaCl solution. The salt immersion test results are depicted in Figure 1.

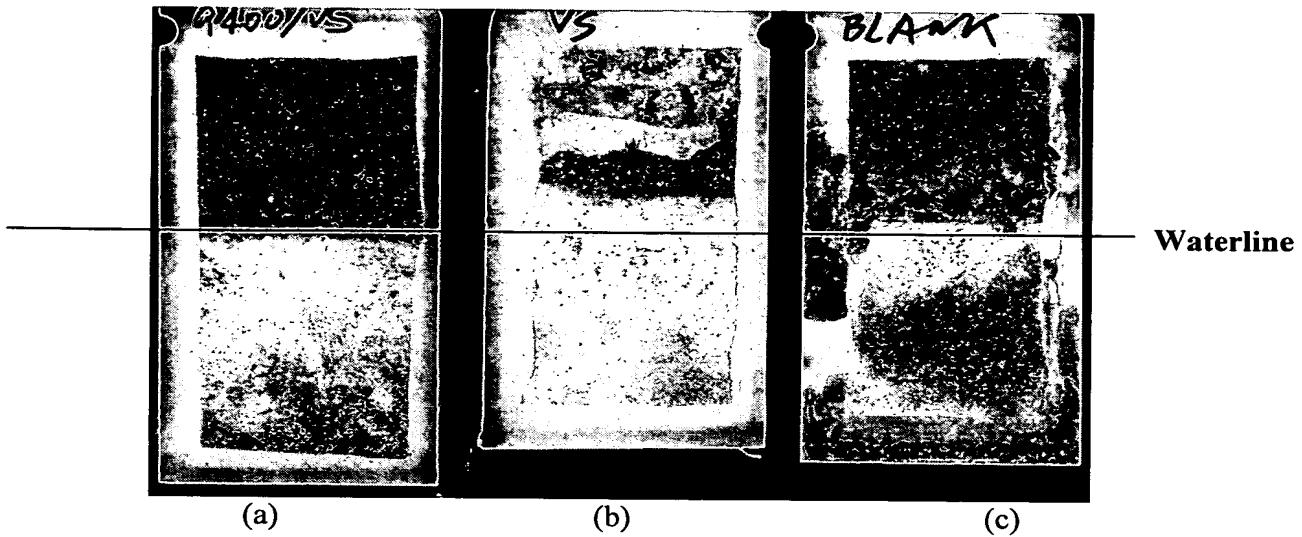


Figure 1. HDG panels after 4 days of immersion in a neutral NaCl solution; (a) mixture of Y9400/VS (5%, 1/9, pH 7.5), (b) VS (5%, pH 6), and (c) untreated.

### Experiment 2

Silane solutions were prepared at 5% for vinyltrimethoxysilane (VS), bis-(trimethoxysilylpropyl) amine ("A-1170") and bis-(trimethoxysilylpropyl) ethylene diamine ("Y9400"). The structure of Y9400 (available from OSi Specialties) is shown on page 10 of the specification. A solvent (ethanol) was used to improve silane solubility, and the ratio of the silane/deionized water/ethanol was 5/65/30 by volume. The solutions were stirred at room temperature in order to hydrolyze the silanes. A first silane solution was mixed at the ratio of 9/1 of VS/Y9400. The first silane mixture was mixed with ethanol and deionized water to yield a 5% silane mixture with a ratio of the silane/deionized water/ethanol of 5/80/15. A second silane solution was mixed at the ratio of 9/1 of VS/A-1170. The second silane mixture was mixed with ethanol and deionized water to yield a 5% silane mixture with a ratio of the silane/deionized water/ethanol of 5/80/15. The pH of the resulting mixtures VS/Y9400 and VS/A-1170 were adjusted using acetic acid to 6. The pH of the 5% vinyl silane solution was 6. Alkaline cleaned hot-dip galvanized steel (HDG, available from ACT Laboratories) panels were dipped into the VS, VS/Y9400 and VS/A-1170 mixtures and then cured at 100 °C for 1.5 hours. Three replicates were prepared for each silane mixture (i.e.,

VS, VS/Y9400 and VS/A-1170). The HDG panels were then subjected to the same stack test as described in the specification. Three HDG panels treated with the same silane mixture were wetted with deionized water and then placed together, i.e. one on top of the other. For a control, three blank HDG panels were wetted with deionized water and then placed together in the same manner as the treated HDG panels. The stacked panels were then placed into a humidity chamber (Temperature=65-70°C, Relative Humidity = ~100%) for 8 days. This stack test employed more aggressive conditions, namely significantly higher temperature than the stack test performed in the specification. Visual inspections were carried out daily. The wet stack test results are provided in Table 1.

Silane	Curing condition	White rust percentage (%)						
		1 day	2 day	3 day	5 day	6 day	7 day	8 day
VS/A1170 9/1, 5%, pH 5-6	100°C/1.5 hr	0	0	0	0	~10	~10	>10
VS/Y9400 9/1, 5%, pH 5-6	100°C/1.5 hr	0	0	0	0	0	~10	>10
VS 5%, pH 6	100°C/1.5 hr	~10	>10	--	--	--	--	--
Blank HDG	---	>10	--	--	--	--	--	--

Table 1. Wet stack test of silane treated HDG panels

### Experiment 3

The stability of the solutions prepared in Experiment 2 were evaluated (i.e., VS, VS/A-1170 and VS/Y9400). A visual inspection of the solutions was carried out at 2 hours, 6 hours and 24 hours after preparation of the solutions. The results of the solution stability test are provided in Table 2.

Silane	Minimum amount of ethanol	Solution stability		
		2 hr	6hr	24 hr
Vinylmethoxysilane (VS), 5%, nat. pH 6	30%	Hazy	Hazy	Hazy+white precipitates
VS/A1170 9/1, 5%, pH 5-6*	15%	Clear	Clear	Hazy

VS/Y9400 9/1, 5%, pH 5-6*	15%	Clear	Clear	Hazy
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\* adjusted by Acetic Acid

Table 2. Solution stability of VS, VS/Y9400 and VS/A1170

4. The experiments described above further demonstrate that the addition of one or more bis-silyl aminosilanes to a vinyl silane solution significantly improves the storage stability of the solution and significantly improves the corrosion protection provided by the solution. The shelf life of the vinylmethoxysilane (VS) solution was prolonged by the addition of a small amount of A1170 or Y9400. In addition, after treatment of the HDG panels with the silane mixtures of the present invention, the salt immersion test of Experiment 1 demonstrates that the addition of even a small amount of a bis-silyl diaminosilane (Y9400) to a vinyl silane solution significantly and surprisingly improves corrosion protection. Moreover, the results of the wet stack test of Experiment 2 further demonstrate that the vinyl silane mixtures of the present invention (VS/A1170 & VS/Y9400) greatly improve protection of HDG panels from white rusting as compared to VS solution alone. The surprising improvements exhibited by the present compositions are not disclosed or rendered obvious by the prior art references cited in the Official Action dated March 19, 2002.

5. As one skilled in the art will appreciate, the showings of the unexpected results of the present experiments and the examples detailed in the present specification, i.e. vinyl silane mixtures containing bis-(trimethoxysilylpropyl) ethylene diamine or bis-(trimethoxysilylpropyl) amine, are commensurate in scope with the claimed invention. In particular, given that the experiments described herein and in the specification clearly show unexpected results for both a bis-silyl "mono"amine (A-1170) and a bis-silyl diaminosilane (Y9400), one skilled in the art would conclude that other bis-silyl aminosilanes encompassed by the claims would provide similar improvements in corrosion protection and solution solubility. By way of example, one skilled in the art would conclude that using a bis-

(triethoxysilylpropyl) amine (see page 10 of specification) in place of A-1170 in the silane mixture would lead to the same surprising results since the ethoxy moieties of this bis-silyl amine would be hydrolyzed to hydrogen moieties in the same fashion as the methoxy moieties of A-1170.

6. Dr. Wim J. van Ooij further declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Respectfully submitted,



Dr. Wim J. van Ooij

9/26/02

Date

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